Thesis

THE ULTRASONIC PRODUCTION OF CAVITATION IN LIQUIDS

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Thesis

THE ULTRASONIC PRODUCTION OF CAVITATION IN LIQUIDS

by

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STATEMENT OF THE PROBLEM

The phenomena of cavitation, produced in liquids by an ultrasonic beam, was observed nearly a quarter of a century ago. It was noticed that many previously unobserved effects accompanied the phenomena; the efficiency and transmitting power of the transducer as well as the physical and chemical properties of the transmitting medium were radically altered. Many of the other physical and chemical phenomena produced by ultrasonic waves were found in some way to be connected with cavitation. The existing theories of the basic structure of liquids were soon found to yield information which was not correlated by experimental data.

The purpose of this paper is to make a comprehensive survey of the phenomena of ultrasonic cavitation in liquids. For better understanding of the problem a brief discussion of the general properties and characteristics of ultrasonic waves, as well as a short history of the development of the science of ultrasonics, has been included. The more recent methods of production of high frequency sound waves, along with the experimental methods and equipment employed in the study of cavitation has also been briefly described.

It is the main purpose of this study (1) to review the present theories of the ultrasonic production of cavitation

in liquids; (2) to evaluate the theories in light of the experimental methods and results; (3) to analyze the properties of liquids in an ultrasonic field; (4) to summarize the more tenable theories of cavitation in view of present day researches in the field of ultrasonics.

Although it has been impossible to thoroughly review the voluminous literature written about ultrasonic phenomena, an attempt has been made to include those which seemed most relevant to the immediate problem.

DEFINITION OF TERMS

In general the terminology of ultrasonics and ordinary sonic phenomena is the same; since ultrasonics is but a special branch of acoustics. In order to avoid confusion, the definition of a few specialized terms is here included.

Ultrasonics. The term "ultrasonic" will be used to denote those frequencies which are beyond the limits of human audibility. These will include all frequencies greater than 20,000 cycles per second (20 kc). The term "supersonic" will be reserved for phenomena that occurs at a speed greater than the speed of sound in air. It will be noticed, that in the literature both terms are used interchangeably.

Cavitation. When an ultrasonic beam of high enough intensity passes through a liquid, bubbles or cavities are formed at regularly defined points in the liquid. This phenomena has been called cavitation. In this paper the term "cavitation" will be used to describe both the formation of gas bubbles, as well as the formation of natural holes or cavities in the liquid.

Transducer. Any element which can be mechanically or electrically excited to produce or receive ultrasonic vibrations.

Group Velocity. This terms applies to the velocity with which the envelope of a wave is propagated when the wave is amplitude modulated. This is the most important velocity encountered

in ultrasonics. Unless otherwise stated the term velocity will be used to mean group velocity.

Longitudinal Waves. The term longitudinal wave applies to that wave motion where the particle displacement is parallel to the direction of propagation of the wave. Since, as a rule, only longitudinal waves can be transmitted through a liquid medium, these will be the most important in this paper.

LIST OF SYMBOLS USED

1. λ = wavelength
2. c = velocity of a sound wave
3. γ = ratio of specific heats
4. ρ = density
5. ν = frequency
6. u = velocity of vibrating particle
7. p_o = hydrostatic pressure
8. p_v = vapour pressure
9. η = viscosity
10. σ = surface tension
11. K = Boltzmann's constant
12. R = Universal gas constant
13. T = absolute temperature
14. h = Planck's constant

INTRODUCTION TO ULTRASONICS (1)

1. General Properties

Ultrasonic waves are essentially plane waves. Because of their short wavelength, rectilinear propagation is characteristic of ultrasonic waves. Generally, beams are propagated from a plane transducer with very little divergence. In an ordinary quartz plate the energy is located in a cone whose half angle of spread is given by

$$\sin A = 1.2 \frac{\lambda}{D}$$

where

A = half angle of spread

D = distance across transducer face

If the medium through which the beam is propagated is full of small discontinuities the ultrasonic energy may be broken up and reflected and refracted haphazardly. This is an important characteristic in considering cavitation in liquids. In general it is usual to assume that the ultrasonic vibrations take place too quickly for heat to exchange. Hence we may write an expression for the velocity of ultrasonic waves in liquids as

$$C = \left(\frac{\gamma}{\rho \beta_{is}}\right)^{1/2} = \left(\frac{1}{\rho \beta_{ad}}\right)^{1/2}$$

(1) B. Carlin, Ultrasonics, McGraw Hill, New York (1949)

where

β is = isothermal compressibility

β ad = adiabatic compressibility

The dissipation of energy in an ultrasonic system is due primarily to the viscosity or damping of the medium. There is also some energy loss due to heat conduction and radiation between compression and rarefaction sections.

2. Range of Frequencies and Wavelengths

From the familiar relationship between frequency, ν, wavelength, λ and velocity, c

$$c = \lambda \nu$$

it is possible to calculate the frequency and wavelength ranges in gases, liquids, and solids if the velocity in that medium is known. Bergmann(2) gives for ultrasonic waves in air (velocity of sound c = 330 m/s) a wavelength range of 1.6 cm. to 0.6×10^{-4} cm., in liquids ($c \simeq 1200$ m/s) a range of 6 cm. to 2.4×10^{-4} cm., and in solids ($c \simeq 4000$ m/s) a range from 20 cm. to 8×10^{-4} cm.

3. Generation of Ultrasonic Waves

The method of generating high frequency sound waves depends upon the power output necessary and the frequency range required. The greatest amount of energy is commonly produced in ultrasonics by systems designed for agitation and cavitation experiments. Energy outputs of 10 watts/cm.^{2}

(2) L. Bergmann, Der Ultraschall, translated by H. S. Hatfield, Ultrasonics and their Scientific and Technical Applications, John Wiley and Sons, New York (1942)

have been reported.[3] The most common way of producing ultrasonic vibrations is by a magnetostriction rod or piezoelectric crystal transducer. The former method is used when frequencies up to 100 kc. are needed while the latter method is used for those frequencies in the range from 100 kc. to 15 mc. In recent years these two methods have been used to the exclusion of others in most cases.

4. Detection of Ultrasonic Waves

When an ultrasonic beam is projected against a boundary it produces two distinct effects. First, there is an alternating pressure at the frequency of wave propagation, and secondly, direct pressure due to the radiation. From any text on acoustics it is shown that the alternating pressure is determined by

$$J = \frac{\rho c}{2}(\omega A)^2 = \frac{P_{\mu}}{2}$$

where

J = intensity/cm.^{2}-sec.
A = maximum amplitude of the vibrating particles
P = sound pressure
C = velocity of sound wave of frequency W/2π

We see that the measurement of sound intensity is possible only if one of the magnitudes A, μ, or P can be measured or alternatively, if both μ and P can be measured. In the ultrasonic range it is impossible to accomplish this with mechanical receivers, since these have dimensions too great compared

(3) B. Carlin, Ref. 1

with the sound waves. However, since we are dealing here with high frequencies we have also comparatively high intensities and are enabled to make use for measurement, of the second effect mentioned above, the sound radiation pressure. This unidirectional pressure finds its theoretical explanation in the nonlinearity of the wave equation when account is taken of the quadratic terms. Sound radiation pressure, S, is given by the relationship

$$S = \frac{1}{2}(\gamma + 1)\frac{J}{c}$$

The measurement of sound radiation pressure is usually made by radiometers.

5. General Theory of Ultrasonic Wave Motion

From any text on elasticity,(4) we have for the general equation of the displacement of a particle in any medium in a periodic force field

$$(\lambda + G)\frac{\partial e}{\partial q_1} + G\nabla^2 u - \rho\frac{\partial^2 u}{\partial t^2} = 0$$

where

u = displacement of the particle in the q_1 direction
λ = Lame's constant
G = modulus of rigidity
e = volume expansion
ρ = density of medium

$$\nabla^2 = \frac{\partial^2}{\partial q_1^2} + \frac{\partial^2}{\partial q_2^2} + \frac{\partial^2}{\partial q_3^2} + \cdots$$

This is a set of differential equations of motion representing types of displacement in which the medium obeys Hooke's

(4) S. Timoshenko, Theory of Elasticity, McGraw Hill, New York (1936)

law and remains in a static state as a whole, i.e., the only forces acting are internal stresses and shears and those due to particle acceleration forces.

The above represents a set of theoretical equations of motion subject to physical and boundary conditions. Since longitudinal waves are the only type of ultrasonic waves possible in liquids, it is possible to assume physical conditions and derive the general equation of motion of the particles of an ideal liquid for such waves. A longitudinal wave has been defined as one in which the particle velocity is restricted to a motion in the direction of the wave propagation. Such waves are dilatational and irrotational; i.e., there is a change in the body volume without rotation. Now since we have the particle displacement u as the only motion, then the volume expansion in the q_1 direction becomes

$$\frac{\partial e}{\partial q_1} = \frac{\partial}{\partial q_1}\frac{\partial e}{\partial q_1} = \frac{\partial^2 e}{\partial q_1^2}$$

assuming e as a function of u alone. Also, since the motion is restricted in the direction of propagation, if we take this as q_1, then since

$$\nabla^2 = \frac{\partial^2}{\partial q_1^2} + \frac{\partial^2}{\partial q_2^2} + \frac{\partial^2}{\partial q_3^2} + \cdots$$

we have

$$\nabla^2 u = \frac{\partial^2 u}{\partial q_1^2}$$

Hence our general equation of motion, essentially the wave

equation, becomes

$$(\lambda + 2G)\frac{\partial^2 u}{\partial x_1^2} = \rho \frac{\partial^2 u}{\partial t^2}$$

In actuality, there is always some shear stress since the edges of the path cannot move back and forth quickly.

Solutions of the above differential equation, with the application of further physical and boundary conditions, will describe, theoretically, an ultrasonic wave field.

6. History of Ultrasonics

The application of ultrasonics in the solution of industrial problems has gained much popularity in the last decade. Although phenomena produced by high frequency sound waves was observed over half a century ago, the science of ultrasonics was slow in its development. This was due primarily to inadequate equipment and lack of knowledge of the basic structure of matter.

Prior to the first world war ultrasonic waves were produced by electric sparks or some mechanical or thermal vibrator. Langevin, in his work on under water signalling, was the first to apply the piezoelectric effect to the generation of ultrasonic waves. This provided the first dependable source of ultrasonic vibrations of controllable frequency and intensity. Using quartz crystal he applied the principle of the piezoelectric effect discovered by Pierre and Jacques Curie in 1880 to produce high frequency vibrations for the purpose

of underwater signalling and submarine detection.(5) Langevin, during his experiments, noticed the lethal effects of ultrasonic waves of high intensity upon small marine life.(6) This was perhaps the first time that the cavitation effects of ultrasonic waves was mentioned. Little was done at the time to further investigate and explain its effects. The fact that more power was made available, if the quartz transducer was vibrating at its resonant frequency, was also discovered by Langevin. In 1922 Cady boosted the advance of ultrasonic research when, as a result of his studies of piezoelectric resonators, he came forth and developed the quartz frequency stabilizer. Three years later, Pierce gave new impetus to research by utilizing a vacuum tube circuit containing a piezoelectric plate for measurement of ultrasonic velocity in carbon dioxide.

In the period 1925-1927 Boyle, and his colleagues in Canada, made a comprehensive study of the properties of these waves; their velocity, reflection, refraction, interference and diffraction in liquids was investigated. It was during the course of these experiments that cavitation in liquids produced by an ultrasonic beam was first investigated and reported.(7) (8) (9)

(5) J. C. Hubbard, Journ. Acoust. Soc. Am.,4,99 (1932)
(6) B. Carlin, Ref. 1
(7) R. W. Boyle and G. B. Taylor, Phys. Rev., (11),27,518 (1926)
(8) G. B. Taylor and D.O. Sproule, Roy.Soc.Can.Trans., (III), 23,91 (1929)
(9) R. W. Boyle, et al, Roy.Soc.Can.Trans.,(III), 23,187 (1929)

The phenomena was observed for ultrasonic waves where the energy was concentrated in the form of a narrow angle beam. The maximum sound energy which could be transmitted through a liquid medium was found to be limited by cavitation. If the hydrostatic pressure p_o is considered as the only preventive of disruption in the medium, then the maximum energy transmitable per square centimeter is given by

$$E_{max}/cm^2 = p_o^2/2\rho c$$

Experiments showed that cavitation is more readily produced in highly volatile liquids, hence it was suggested that the vapour pressure, p_v, be taken into consideration. This gives for the maximum energy transmitable

$$E_{max}/cm^2 = (p_o - p_v)^2/2\rho c$$

Experiments showed that the cavitation point was far below that given by the above equation. It is now known that other factors besides those listed above limit the power output.

Study of the effects of intense ultrasonic radiation began in 1927 with Wood and Loomis[10] who carried out extensive experiments using a specially designed vacuum tube generator capable of 2000 watt output.[11] Shortly thereafter it became apparent that emulsification, the destruction of bacteria, the freeing of antibodies from bacterial cells,

(10) R.W.Wood and A.L.Loomis, Phys. Rev., 11, 29 (1927)
(11) R.W.Wood and A.L.Loomis, Phil.Mag., 4, 417 (1927)

the acceleration of chemical reactions, the production of luminescence and several other phenomena, observed in an intense ultrasonic wave field, were dependent for their existence upon cavitation. It was also observed that in degassed liquids, or in liquids under high hydrostatic pressures, most of these effects either disappeared or proceeded very feebly.

In 1932 Debye and Sears[12] performed light scattering experiments with ultrasonic waves in which they observed diffraction patterns. There is a possibility that this phenomena is due to cavitation. In the early 1930's, physicists and chemists used ultrasonics to further study specific heats of gases with respect to the relative parts played by vibrational and rotational frequencies of molecules at high temperature. This was approached through a study of absorption and velocity determination in gases as well as in liquids. Much research along these lines is still in progress.

Further progress in the investigation of ultrasonic cavitation had to wait for a new theory of the physical structure of liquids. H. Eyring in 1936 advanced some new ideas on liquid structure which was extensively developed by R. Fürth in 1941. This theory will be discussed more thoroughly later.

The greatest avenues of further development of ultrasonic research lie in the fields of metallurgy and biophysics.

(12) P. Debye and F.W.Sears, Proc.Natl. Acad. Sci.U.S., 18,410 (1932)

Before any real progress can evolve in these fields, a concise mathematical and physical analysis of ultrasonic cavitation is necessary. A great deal remains to be accomplished

EXPERIMENTAL PROCEDURES IN THE INVESTIGATION OF ULTRASONIC CAVITATION[13]

1. Apparatus

The following is a brief account of the experimental procedure and apparatus used by Briggs[13] and his colleagues during a series of experiments designed to test the cavitation strength of various liquids. A sectional view of the test chamber and crystal units used for studying cavitation is shown in Fig. 1*. The major faces of the crystals were coated with gold and cemented together in groups of four. This crystal group was then cemented to a quarter wave resonator block which made a half wave unit with the node at the interface between the crystal and resonator block. This unit fitted into a cup faced with rho-c rubber (a rubber having the same density and dilation velocity as water), into which was placed the liquid to be tested.

The radiator radiated waves through the liquid, rubber and standpipe to a second crystal unit where they were picked up and generated into an electrical replica which was put on a cathode ray tube. In order to avoid cavitation in the standpipe, it was filled with degassed castor oil. By the use of a vacuum pump and a rotary oil pump, the liquid to be

(13) H.B.Briggs, et al, Journ. Acoust.Soc.Am., 19,664 (1947)

* All figures and tables mentioned may be found in Ref. 13

tested for cavitation resistance was also thoroughly degassed.

A schematic diagram of the power source and circuits used for testing liquids is shown in Fig. 2. The power input, W, to the quartz crystal projector was calculated from the voltage V and resonance resistance R of the projector using the relationship

$$W = V^2/R$$

Two power supplies were available; one a 75 watt supply used for either pulsing or steady power, the other a 1000 watt supply used only for pulsing power.

For steady power input, the voltage across the crystal terminals was read off directly on a vacuum tube voltmeter, while the minimum resistance was taken as the resonance resistance. For pulsing tests the voltage applied to the projector was determined from the amplitude of the oscilloscope deflection. The oscilloscope also served to measure the relative magnitude of response picked up by the receiver for varying inputs. It was, therefore, possible to draw input-cutput curves as shown in Fig. 3 for non-degassed kerosene. Wave form or pulse form, as shown by the oscilloscope, indicated when cavitation was present.

2. Procedure

Assuming the radiator to be 100 percent efficient,

peak pressures can be calculated approximately by the formula

$$W = \frac{1}{2} p_m^2 A/\rho c$$

or

$$p_m = [(2\rho c/A)W]^{1/2}$$

where

p_m = peak pressure in dynes/cm^2
A = radiating area in cm^2
W = power output in ergs/sec

As an example, we examine the input-output curve of Fig. 3. The relationship between input and output is linear up to 800 volts (cavitation point) after which it bends and rises very slowly. It was found that the resonance resistance was 2×10^5 ohms. With a projector surface area of 6.45 cm^2 we get for the power output per unit area of projector

$$W/cm^2 = V^2/RA = 0.5 \text{ watts/cm}^2$$

If the density of non-degassed kerosene is taken as 0.81 gm/cm^2 and the velocity of the wave in the liquid as 1.324×10^5 cm/sec, we get for the maximum pressure p_m a value a little larger than 10^6 dynes/cm^2, which is roughly atmospheric pressure. Hence, it would seem safe to conclude in this case that a light undegassed liquid is held together principally by atmospheric pressure and does not exhibit any cohesive force.

3. Results of the Experiments

For a better understanding of the process of cavitation,

Briggs[14] made measurements on liquids having wide ranges of viscosity, vapor pressure and chemical structure. Table I lists some of the properties of these liquids. It is to be noted that the vapour pressures are not given specifically but rather as lying within certain ranges.

The first series of tests were made on the effects of dissolved gases on cavitation voltage. By varying the pressure on the liquid the effect of hydrostatic pressure was determined. Fig. 4 shows the voltage across the crystal at which cavitation occurred as a function of the ambient pressure for degassed kerosene. The resultant curve is a straight line which when extended cuts the axis at a negative pressure of 60 cm. of mercury. Hence degassed kerosene will stand a negative pressure of nearly one atmosphere before cavitation occurs. We can also conclude that the cavitation pressure is the sum of the cohesive pressure (tensile strength) plus the hydrostatic pressure.

Measurements which were made on the cohesive pressure of the liquids used are given in the last column of Table I. Pc is the peak acoustic pressure at which cavitation begins at one atmosphere hydrostatic pressure in the liquid. The cohesive pressure p_c is then

$$p_c = Pc - 1$$

The value of p_c depends to a large extent on the previous

(14) H. B. Briggs, et al, Ref. 13

history of the liquid. A cavitated liquid is "weakened" and will require some time to return to its earlier state; if the experiment is repeated the liquid will cavitate at a lower acoustic pressure.

It was found that the only quantity consistently related to the cohesive pressure is the viscosity. Fig.5 shows cohesive pressure as a function of viscosity, the latter being plotted on a logarithmic scale. Briggs found the equation of this curve, which is a straight line within experimental error, to be

$$\ln(\eta/\eta_0) = 3.05\, p_c$$

where

η = viscosity in poises
η_0 = 0.0013 poise
p_c = cohesive pressure in atmospheres

From the above equation it is seen that as η approaches η_0 the cohesive pressure approaches zero. Hence when the viscosity approaches 0.0013 poise the liquid is held together only by the static pressure of the atmosphere. The theoretical interpretation of the cavitation point of a liquid as a function of viscosity will be given when Eyring's theory of viscosity is considered.

PRODUCTION AND PROPERTIES OF CAVITIES IN LIQUIDS

1. Production

When a piezoelectric quartz crystal is immersed in a liquid and made to vibrate at ultrasonic frequencies, bubbles or cavities are observed in the liquid if conditions are favorable. The simplest arrangement is probably found in the ultrasonic interferometer, a description of which is found in Colby.[15] If a solid reflecting surface is placed an integral number of high wave lengths from the transducer, a stationary wave is set up in the liquid with a displacement node at the face of the transducer and also at the reflecting wall. Since in a stationary wave the displacement wave and compression wave are ninety degrees out of phase, we find pressure loops and displacement nodes coinciding. Cavities are formed during the negative half of the pressure cycle, and are observed to collect at pressure loops of the stationary wave pattern. Hence, we find cavities present at the faces of the transducer and reflector and also at distances of $\lambda/2$, where λ is the wavelength, from the transducer. It will be sufficient at this point to explain the formation of these cavities as a "tearing apart" of the molecules of the liquid, which leaves empty spaces behind.

(15) M. Y. Colby, Sound Waves and Acoustics, Henry Holt and Co., New York (1938)

2. Properties

According to Sollner[16] these cavities may be made visible by irradiating long layers of completely degassed liquid with an ultrasonic beam of high intensity. Due to the nearly sinusoidal pressure distribution in the liquid, we have zones of greater density, therefore less transparency to light. These zones disappear, when the beam is shut off, without leaving bubbles behind. An extension of this phenomena is used in the design of the ultrasonic diffraction "grating".

An ingenious method of determining experimentally the loci of equal phase in an ultrasonic compressional wave beam without utilizing stationary wave patterns, has been devised by Labow.[17] The electric signal generated by the piezoelectric action of a Rochelle salt microphone, which has the same phase as the compression wave in the liquid, is combined with a small electric signal of the same frequency having the phase of the transducer voltage. The result of this superposition is a sinusoidal spatial variation of the signal generated by the microphone as the latter is moved in the direction of the beam. The wave front is traced by following a maximum of this sinusoidal variation as the

(16) K. Söllner, Trans. Faraday Soc. 32, 1537 (1936)
(17) L.W.Labow, Journ. Acoust. Soc. Am., 17,19 (1945)

microphone is made to move perpendicular to the direction of propagation of the beam. Hence it is possible to determine where cavitation should take place if it is not visible. However, with proper power input, frequency and a sufficiently gassed liquid, the vapor dissolved in the liquid soon fills up the holes caused by the tearing apart of the molecules, forming gas bubbles which grow until sufficiently big to be visible.

These spherical bubbles of gas pulsate radially under the influence of the alternating sound pressure.(18) The pulsating bubble has potential energy from the elasticity of the gas and kinetic energy arising from the movement of the liquid in the vicinity. We have resonance occurring at that frequency when the mean potential and kinetic energies are equal. Smith has shown that the lowest resonance frequency of a bubble formed by cavitation is given by

$$f = \frac{1}{2\pi r}\left[\frac{3\gamma(P_0 + 2\sigma/r)}{\rho}\right]^{1/2}$$

where

f = resonant frequency
r = radius of the bubble
σ = surface tension of liquid gas interface

For water under standard conditions a bubble of 0.033 mm. radius has a resonant frequency of 100 kc. These resonant

(18) F.D.Smith, Phil.Mag., 19, 1147 (1935)

bubbles are characterized by violent activity and appear to the eye as a luminous streak on account of the persistence of vision. Larger and smaller bubbles are static, since due to a change in radius the bubbles no longer resonate to the frequency of the applied field. "The irregular motion of the bubbles in such a field may possibly be explained as due to Bjerknes forces set up between the bubbles and the sound field with attraction or repulsion between the bubbles depending on the relative phase of their vibrations. The motions of the bubbles bear some similarity to the behavior of charged particles in a varying electrostatic field."(19) Richards(20) has observed that cavitation is accompanied by severe electrical disturbances in liquids. Over distances molecular in size these may be great enough to produce ionization, combination, etc. which may explain the behavior of these gas bubbles.

Some of the properties of these bubbles, formed by cavitation, are brought out if we follow the derivation for the frequency of a resonant bubble given above by Smith. Bubbles are small in comparison with the wavelength, hence the alternating sound pressure acts uniformly over the surface of the bubble and they pulsate in the simplest radial mode. Each bubble has a restoring force, inertia and damping and pulsates as a resonant mechanical system which has one degree of freedom.

(19) H.B.Briggs, etal, Journ.Acoust.Soc.Am., 19,664 (1947)
(20) W.T.Richards, Journ.Appl.Phys., 9, 298 (1938)

The restoring force per unit area which arises from the elasticity of the gas in the bubble is

$$\Gamma = \frac{3\gamma}{r}\left(p_0 + \frac{2\sigma}{r}\right)$$

We will assume here that the compressions and expansions of the gas in the bubbles take place too fast for any heat transfer, hence an adiabatic process. In the last expression Γ is the restoring force per unit area, while γ, r, p_o and σ have the same meaning as before.

The effective mass per unit area, α, of a pulsating bubble arises from the kinetic energy of the liquid moved radially by the pulsations[21] and is given by

$$\alpha = \rho r\left(1 + k^2 r^2\right)$$

where

$$k = \frac{2\pi}{r}$$

The radiation resistance or damping factor per unit area β arises from the scattering of the sound by the bubble[22] and is given by

$$\beta = \rho c^2 k^2 r^2 / \left(1 + k^2 r^2\right)$$

where

c = velocity of sound in the liquid

(21) R.W.Wood and A.L.Loomis, Phil.Mag., 4,417 (1927)
(22) J.B.Crandall, The Theory of Vibrating Systems and Sound D. Van Nostrand and Co., New York (1926)

Now, if we call p equal to the instantaneous alternating pressure in the liquid in the absence of a bubble, then the equation of motion of the bubble is given by

$$p = \alpha \frac{d^2\xi}{dt^2} + \beta \frac{d\xi}{dt} + \Gamma \xi$$

where ξ is the displacement of the surface of the bubble from its equilibrium position. This is analogous to electrical and mechanical forced oscillatory systems.

From the theory of forced oscillations we have resonance occurring when

$$\alpha \frac{d^2\xi}{dt^2} + \Gamma \xi = 0$$

or when

$$p = \beta \frac{d\xi}{dt}$$

That is, resonance will occur when we have a frequency f given by

$$f = \frac{1}{2\pi} \left(\frac{\Gamma}{\alpha} \right)^{1/2}$$

If we substitute for Γ and α we have

$$f = \frac{1}{2\pi r} \left[\frac{3\gamma (p_0 + 2\sigma/r)}{\rho (1 + k^2 r^2)} \right]^{1/2}$$

which gives the resonant frequency. In his derivation for f Smith assumed that for water $k^2r^2 \ll 1$, hence his formula follows. It may well be that these resonant bubbles are the cause of the destructive phenomena associated with cavitation.

STRUCTURE OF LIQUIDS AND THEIR PROPERTIES IN AN ULTRASONIC FIELD

Fürth[23] has suggested a new theory of liquid structure in which holes in a continuum are treated by the methods of statistical mechanics.[24] This is exactly the reverse of the treatment of solid particles in empty space encountered in kinetic theory. These holes are assumed to contain vapour at the vapour pressure corresponding to the temperature. This differs from Eyring's[25] theory of holes in that Eyring assumes the existence of natural holes in a liquid, which, when in the equilibrium state, are devoid of vapour.

Fürth's treatment assumes the existence of a known metastable state in which the temperature exceeds the saturation value corresponding to the pressure; the vapour pressure exceeding the hydrostatic pressure. For the limit of this metastable state he obtains the following equation

$$p_v - p_0 = \frac{1.3\,\sigma^{3/2}}{(KT)^{1/2}}$$

where

T = temperature

(23) R.Furth, Proc. Camb. Phil. Soc., 37,252 (1941)
(24) R.S.Silver, Nature, 150,605 (1942)
(25) H.Eyring, J.Chem., Phys. 4,283 (1936)

For example, for water at normal temperature we get $p_v - p_o = 8 \times 10^4$ lbs/in^2.

Now if p_o is considered atmospheric pressure then the excess of vapour pressure demands a high temperature before it can be realized, and in this case has little meaning since the pressure is far above the critical breaking pressure of the liquid. In reality the right hand side of the above equation needs to be altered to allow for different values of the temperature. As the equation stands above the two sides become equated for water at a temperature of about 337°C when the vapour pressure is in excess of 2000 lbs/in^2.

The superheating of water has never been attained to this degree, due to the presence of particles and dissolved gasses. The highest reached is about 180°C for water under artificial conditions.

There is another way of interpreting this equation. It is usually assumed that the hydrostatic pressure is equal to or less than zero, whence at a given temperature $p_v - p_o$ is less than or equal to the vapour pressure. However, if the hydrostatic pressure is taken less than zero then the relationship between $p_v - p_o$ and the vapour pressure given above is no longer true. From the above data it appears that the equation defining the limit of the metastable state would be satisfied if the hydrostatic pressure were of the order of 8×10^4 lbs/in^2,

since the vapour pressure at standard temperature is low enough to be negligible. From this we might infer that the liquid should theoretically have a tensile strength up to the limit of the hydrostatic pressure. However, particles and dissolved gases in the liquid tend to lower the limit far below the expected value. From elementary considerations it can be shown that in the case of water, for example, the tensile strength is negligible. This seems to be borne out by experimental facts as was shown earlier by Briggs for undegassed kerosene. If we employ the equation for equilibrium of a bubble of radius r, it is found that the tensile strength limit lies between $2\sigma/r$ and $4\sigma/3r$. Bubbles with radii as small as 10^{-4}cm. would make this limit of the order of about one atmosphere. Under ordinary circumstances evaporation nuclei of much greater radius exist in water, decreasing still further the tensile strength.

Silver[26] suggests the following interpretation to the seeming fallacy in Fürth's equation for the limit of a metastable state of a liquid in which vapour filled holes are assumed. While evaporation nuclei may exist in a liquid, evaporation will not take place unless there exists a suitable temperature distribution formed to allow the conduction of heat into these nuclei. Also, this must occur at a limited rate governed by the distribution of the temperature and the surface area of these nuclei. Hence, we may deduce

(26) R.S.Silver, Nature 150, 605 (1942)

that the tensile stress above the limit imposed by the nuclei may persist for a short time. The value attained depends on the relative times for evaporation and also for the growth of the stress. The suggestion is that the ultimate limit corresponding to the hydrostatic pressure derived from Fürth could be carried for an infinitesimal time interval and still be transmissable as a tensile wave of rectangular wave front. For example, a very steep tensile wave front is to be expected for water.

The problem then is to find a peak tension before the liquid ruptures[27]. Consider a liquid saturated with gas. If some of this liquid is enclosed in a container and tension is applied, we have supersaturation due to the reduced pressure. A bubble of gas would form and a new equilibrium state would be reached. Yet some time is necessary for diffusion to the weakest boundary to form the initial bubble and the saturated liquid would stand a momentary tension. However, if the liquid is completely degassed the pressure can still be reduced below its own vapour pressure without rupture, since completely degassed liquids have a tensile strength which permits sound pressure higher than the hydrostatic pressure to be transmitted.

If we consider Van der Waals equation of state which

(27) H.B.Briggs, et al., Ref. 13

holds approximately for liquids

$$(p + a/v^2)(v - b) = RT$$

we notice that as the pressure p is varied the volume v changes but little, hence the term a/v^2 may be considered a constant. It is in the form of an external force that holds the volume of the liquid constant although its shape may change. The magnitude of the internal tension may be estimated from the compressibility of liquids which is of the order of about $\Delta v/v = 10^{-4}$ per atmosphere pressure. The inward pressure a/v^2 is of the order of 10^4 atmospheres, and application of ordinary pressures externally changes the volume of the liquid but little. Attempts to measure the static breaking strength of liquids give erratic values far below the theoretical values.

Vincent[28][29] made a study of the stability of a gas bubble in a liquid. Any gas bubble should be unstable since it is buoyant and will tend to rise and enlarge in a liquid. A temporary stability may be established if the pressure inside the bubble is greater than the hydrostatic pressure. It is clear that the smaller the bubble the greater the internal pressure must be for stability. It is fairly simple to calculate what the radius of the largest bubble can be if we let

(28) R.S.Vincent, Proc.Phys.Soc.London 53,126 (1941)
(29) R.S.Vincent, Proc.Phys.Soc.London 55,41 (1943)

r = radius of the bubble
and p = internal pressure.

Clearly the force of pressure on an equatorial cross section of bubble is

$$F_e = (p - p_o)\pi r^2$$

and this is balanced (for stability) by the force of surface tension along the periphery

$$Fp = 2\pi r\sigma$$

The total outward force is then

$$F = F_e - Fp$$

or

$$F = (p - p_o)\pi r^2 - 2\pi r\sigma$$

Now if the bubble is to be stable the total outward force must equal zero and hence

$$r = \frac{2\sigma}{(p - p_o)}$$

For instance, Vincent found that the highest breaking strength in oil is about 7.8 atmospheres. We can now calculate the radius of the largest stable bubble present. The surface tension for oil is about 30 dynes/cm. and the hydrostatic pressure is -7.8×10^6 dynes/cm^2. Assuming that the hydrostatic pressure in the bubble is negligible, the bubble which became unstable and caused the break was no greater than 7.7×10^{-4}cm. just before the break.

A bubble of such dimensions has very little buoyancy

and is eliminated from the liquid with difficulty even if near the surface. If as has been assumed by Furth and Eyring, mobile holes exist in a liquid, then the process of rupture consists in one of the larger holes becoming unstable under the tension applied to the liquid and the gas that appears arrives in the enlarged cavity by subsequent diffusion from the liquid.

It can be concluded then that the formation of cavities of unstable size in a degassed liquid has to proceed unaided by gas pressure. At a very early stage of development of the cavity, externally applied tension on the liquid fails to be supported by any internal tension; the cavity then fails to be an abnormal point in the liquid and becomes a discrete void enclosed by a boundary where the properties of surface tension appear. Now if the external tension is released the cavity should collapse. However, if conditions are not favorable for collapse, a time lag is involved before rupture occurs. This is a normal condition in all the dynamic measurements involved in ultrasonic cavitation experiments made by Briggs[30] and his colleagues.

(30) H.B.Briggs, et al, Ref. 13

HYDRODYNAMIC THEORY OF CAVITATION

1. Introduction

A hydrodynamic theory of cavitation in liquids has been suggested by Kennard[31] for idealized liquids. It is well to keep in mind at this point that the primary effect of the passage of an ultrasonic beam through a liquid is a temperoral and spatial redistribution of the pressure within the liquid. The liquid will cavitate when its pressure sinks to a breaking pressure p_b, then the pressure rises at once to a fixed cavity pressure p_c where $p_b \leq p_c$. Hence p_b is the minimum pressure that a liquid can stand. Certain assumptions need to be made here in order to keep the nature of cavitation from becoming too complex to handle mathematically. "I have been told by every mathematical physicist I know that the analysis of cavitation is a task beyond the ability of present day mathematics."[32]

It will be assumed here that the pressure in the bubbles is constant, the bubbles are small and contain only vapour of the liquid when the cavity pressure equals the vapour pressure, and that the motion is irrotational in a homogeneous liquid of nearly constant density. Hence, the variation of density due to the passage of an ultrasonic beam can be neglected here.

(31) E.H.Kennard, Phys.Rev. 63, 172 (1943)
(32) W.T.Richards, Ref.20

2. Plane Breaking Fronts

The one dimensional equations of motion for acoustic phenomena are given as

$$\frac{\partial u}{\partial t} = -\frac{1}{\rho}\frac{\partial p}{\partial x} \quad ; \quad \frac{\partial p}{\partial t} = -\rho c^2 \frac{\partial u}{\partial x}$$

where

p = excess of pressure above hydrostatic
t = time
ρ = undisturbed density

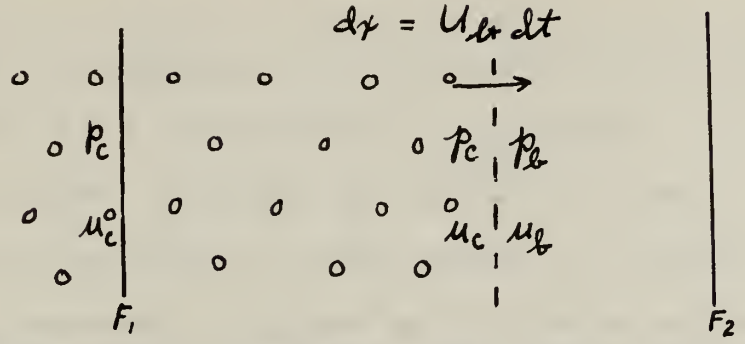
The derivatives in the above equations are assumed to be continuous functions. These equations may be applied generally to the relative motion of the liquid in the neighborhood of any point if uniformly moving axes are employed relative to which the velocity of the liquid near the point is very small.

It has been already asserted that cavitation begins when the pressure sinks to the breaking pressure, hence when

$$p = p_b \text{ then } \frac{\partial p}{\partial t} < 0 \quad ; \quad \frac{\partial u}{\partial x} > 0$$

From the point where p equals p_b a definite breaking front will advance toward both sides; that front advancing toward the right advances into regions where $\frac{\partial p}{\partial x} \geq 0$ while the front advancing to the left advances into regions where $\frac{\partial p}{\partial x} \leq 0$. Just ahead of each breaking front conditions that held at p equals p_b must also hold.

In order to find the rate of advance of a breaking front we notice what happens when the front moves to the right a distance dx in a time dt.



From the above diagram we see that if U_b is the velocity of the plane breaking front, then at the beginning of dt the pressure in the uncavitated liquid is p_b over the left hand face of the layer F_1, while it is $p_b + \left(\frac{\partial p}{\partial x}\right) dx$ over the right hand face of F_2. At the end of dt the pressure over the right hand face of F_2 is also p_b. Then the change in pressure at F_2 during the time interval dt is $-\left(\frac{\partial p}{\partial x}\right) dx$, or since U_b is the speed of the plane breaking front relative to the liquid so that dx equals $U_b dt$, then the change may be expressed as

$$\left(\frac{\partial p}{\partial t}\right) dt \Big|_{x = F_2} = - U_b \left(\frac{\partial p}{\partial x}\right) dt$$

Now since

$$\frac{\partial p}{\partial t} = - \rho c^2 \frac{\partial u}{\partial x}$$

then

$$- U_b \left(\frac{\partial p}{\partial x}\right) dt = - \rho c^2 \left(\frac{\partial u}{\partial x}\right) dt$$

We get then for the velocity of the front

$$U_b = \rho c^2 \left(\frac{\partial u}{\partial t}\right) \Big/ \left(\frac{\partial p}{\partial t}\right)$$

From the diagram it is seen that at the boundary of the front there is a discontinuity in pressure. To find the impulsive effect consider the motion of a layer of liquid dx. On the right hand face F_2 the pressure is nearly equal to the breaking pressure. It sinks to this value as the front comes up to the face, whereas on the left hand face we have acting the steady cavity pressure. The momentum change in the layer dx during the time dt is then given by $(p_c - p_b)dt$ per unit area. If u_b is equal to the particle velocity just ahead of the front and u_c is the particle velocity just behind it, then the velocity changes impulsively from u_b to u_c as the front passes, and since we have $dx = U_b dt$ then

$$(p_c - p_b)\,dt = (u_c - u_b)\,\rho\,dx = (u_c - u_b)\,\rho\,U_b\,dt$$

or

$$u_c = u_b + \frac{(p_c - p_b)}{\rho U_b} = u_b + \frac{(p_c - p_b)}{\rho^2 c^2}\left(\frac{\partial p/\partial t}{\partial u/\partial t}\right)$$

This gives us the particle velocity just behind the plane breaking front. The same can be shown to be true for the breaking front advancing to the left. It can also be shown that the motion of the liquid as given by u_c above is hydrodynamically possible. There are two basic arguments for the

proof of this - one where $p_c = p_b$, the other where $p_c > p_b$. Both of these arguments lead to the same result; namely, that the square of the velocity of the breaking front is equal to or greater than the square of the velocity of the sound wave in the liquid. From this it follows that $U_b = \pm c$, but if this is true for any finite time interval then cavitation does not really occur. We can conclude, then, that a true breaking front always travels faster than sound in the liquid. The conditions which must exist for the propagation of a plane breaking front may be summarized as follows:

$$p = p_b \; ; \left(\frac{\partial u}{\partial x}\right) > 0 \; ; \left|\frac{\partial p}{\partial x}\right| < \rho c \left(\frac{\partial u}{\partial x}\right)$$

and also $\left(\frac{\partial p}{\partial x}\right) \geq 0$ for propagation to the right and $\left(\frac{\partial p}{\partial x}\right) \leq 0$ for propagation to the left.

All of these derivatives are evaluated just ahead of the front since no hydrodynamic influence of the first order can be propagated through a liquid at a speed faster than sound. Once started, a breaking front will continue advancing until it arrives at a point beyond which the necessary conditions stated above are no longer satisfied.

3. The Cavitated Region

In the cavitated region the pressure is uniformly p_c, hence we have $\left(\frac{\partial p}{\partial x}\right) = 0$ and the particle velocity u_c, as

derived above, remains as it was when the breaking front passed by. Now let ϕ equal the fraction of the space occupied by bubbles. Then if $p_o = p_b$, $\phi = 0$ at the trailing edge of the front. But if $p_c > p_b$ then ϕ starts from ϕ_o which is equal to the difference between the space freed by compression from the pressure p_b to the pressure p_c and the space filled by the incoming liquid.

When a front travels to the right, the initial free space in a layer of liquid dx just back of the front is given by

$$\phi_o = [(p_c - p_\ell)/\rho c^2][1 - (c^2/u_\ell^2)]$$

Thereafter we have a change in ϕ at a rate of $\left(\frac{\partial u_c}{\partial x}\right)$. Hence at any time t subsequent to the passage of the front at the time t_b we have

$$\phi = \int_{t_b}^{t} (\partial u_c/\partial x)\, dt + \phi_o$$

This shows that the value of ϕ may vary from point to point in the cavitated region, and even though ϕ_o is small, the value of ϕ may increase to any magnitude.

4. Motion of Plane Breaking Fronts

No influence upon the molecules of a liquid can be propagated with a velocity greater than the velocity of sound in the liquid past a breaking front. Hence the pressure and particle velocity ahead of a front is determined by the

initial and boundary conditions elsewhere. If the boundary of the cavitated region moves with a velocity less than the speed of sound, then cavitation is able to influence the pressure and particle velocity in its neighborhood where the liquid is uncavitated. Any one dimensional disturbance in an unbroken liquid can be resolved into trains of waves going in opposite directions at the same speed. If we let p_1, p_2 and u_1, u_2 denote corresponding pressures and particle velocities respectively, then we can write for the two waves

$$p_1 = \rho c u_1 \qquad p_2 = -\rho c u_2$$

$$p = p_1 + p_2 \qquad u = u_1 + u_2$$

$$p_1 = \tfrac{1}{2}(p + \rho c u) \qquad p_2 = \tfrac{1}{2}(p - \rho c u)$$

If we substitute these into the condition

$$\left(\frac{\partial p}{\partial y}\right) > 0$$

then

$$\left(\frac{\partial p_1}{\partial y}\right) - \left(\frac{\partial p_2}{\partial y}\right) > 0$$

If substituted into the condition

$$\left|\frac{\partial p}{\partial y}\right| < \rho c \left(\frac{\partial u}{\partial y}\right)$$

and both sides are squared, then

$$\left[\left(\frac{\partial p_1}{\partial y}\right) + \left(\frac{\partial p_2}{\partial y}\right)\right]^2 < \left[\left(\frac{\partial p_1}{\partial y}\right) - \left(\frac{\partial p_2}{\partial y}\right)\right]^2$$

or

$$\left(\frac{\partial p_1}{\partial y}\right)\left(\frac{\partial p_2}{\partial y}\right) < 0$$

The first and last of the preceding inequalities can be true if $\left(\frac{\partial p_1}{\partial \gamma}\right)$ is positive and $\left(\frac{\partial p_2}{\partial \gamma}\right)$ is negative. Hence the conditions for the propagation of a plane breaking front, along with the expressions for U_b and u_c, can be written, since $u_b = u$, in the form

$$p_b = p_1 + p_2$$

$$\left(\frac{\partial p_1}{\partial \gamma}\right) > 0$$

and

$$\left(\frac{\partial p_2}{\partial \gamma}\right) < 0$$

From the preceding conditions we can conclude that any wave of tension, such as an ultrasonic wave, can give rise to cavitation only when it is superposed on a wave of decreasing pressure which is travelling in the opposite direction. This is exactly what happens when an ultrasonic stationary wave is set up in a liquid.

5. Plane Resting Boundaries and Closing-Fronts

We shall now consider the behavior of the boundary of a cavitated region when it cannot advance as a plane breaking front. This is the case if a liquid is cavitated ultrasonically. Although a cavitated region will advance as a plane breaking front in a liquid, it is a qualitative fact that the motion of this front ceases when it reaches the nodes of a stationary ultrasonic wave pattern. The boundary

may stand still or it may advance into a cavitated region leaving the liquid behind it uncavitated, in which case the boundary is called a closing-front. Which alternative occurs depends upon the magnitude of

p' = the pressure in the compressional wave train that approaches the boundary in the uncavitated liquid

u_c = particle velocity on the cavitated side of the front boundary

$\frac{\partial u_c}{\partial x}$ = the gradient of the particle velocity

ϕ = the fraction of the space occupied by the cavities

In general, we have three cases of importance

(a) Intrinsic closing-fronts

In this case we have for initial conditions

$$\phi = 0 \quad ; \quad \left(\frac{\partial u_c}{\partial x}\right) < 0$$

and

$$-\left(\frac{\partial u}{\partial x}\right) \geq c \frac{\partial \phi}{\partial x} > 0$$

where x is the distance from the boundary into the cavitated region.

Because of the first two conditions, cavitation disappears in the layers next to the boundary; hence the boundary advances into the cavitated region as a closing-front, leaving the uncavitated liquid behind. This is called an intrinsic closing-front and cannot move through the liquid at velocities greater than c.

(b) Resting boundaries

Here we have the condition

$$2p' \leq p_c + \rho c u_c$$

The pressure is continuous at the boundary and equal to p_c. When the above equation holds but the boundary cannot advance as an intrinsic closing front at a velocity greater than c, then the boundary remains stationary and the incident waves reflected from it are described by the condition that

$$p'' = p_c - p'$$

where p" is equal to the pressure in the receding wave train.

(c) Forced closing-fronts

The initial conditions in this case are

$$2p' > p_c + \rho c u_c \quad ; \quad \phi > 0$$

When the above is true the incident waves are strong enough so that the boundary advances into the cavitated region and causes the bubbles to collapse. This action causes a finite discontinuity of pressure at the boundary if $\phi > 0$. A boundary moving in this fashion is called a forced closing-front. It can be shown that these travel at a speed less than c relative either to the unbroken liquid behind them or to the cavitated liquid in front.

6. Three Dimensional Cavitation

By generalizing from the one dimensional equations stated above, we get for the acoustical equations describing three dimensional phenomena

$$\frac{\partial u}{\partial t} = -\frac{1}{\rho}\nabla p \quad ; \quad \frac{\partial p}{\partial t} = -\rho c^2 \, div \, u$$

where

$$\nabla p = i\frac{\partial p}{\partial x} + j\frac{\partial p}{\partial y} + k\frac{\partial p}{\partial z}$$

i, j, k are unit vectors in the x, y, z directions respectively, and

$$div \, u = \frac{\partial u_x}{\partial x} + \frac{\partial u_y}{\partial y} + \frac{\partial u_z}{\partial z}$$

"Cavitation will begin at a point where a local minimum of pressure sinks to the breaking pressure p_b, and from this point a breaking-front will spread out in the form of a closed surface surrounding the point of initiation and enclosing a cavitated region." (33)

Generalizations from the one dimensional treatment lead to conditions to be satisfied in the uncavitated liquid so that a front may advance, expressable as

$$p = p_b \; ; \; div \, u > 0 \leq \left(\frac{\partial p}{\partial n}\right) < \rho c \, div \, u$$

where n is equal to the distance into the unbroken liquid along the normal to the boundary. Also for U_b, the velocity

(33) E.H.Kennard, p.180, Ref.32

of a front along its normal relative to the unbroken liquid ahead, we get

$$U_b = \rho c^2 \operatorname{div} u \Big/ \left(\frac{\partial p}{\partial n}\right) \geq c$$

By similar generalizations we get for ϕ

$$\phi = \int_{t_0}^{t} \operatorname{div} u_c \, dt + \phi_0$$

In general, great difficulty is encountered when a complete theory of three dimensional cavitation is attempted hydrodynamically. Consider for example the special case of spherical waves.

Let $p_1 = \frac{1}{r} f(ct - r)$ be the pressure in a train of waves from a fixed center, where r is a parameter from that center. Then let these waves be superposed upon other waves expressed as $p_2(x,y,z,t)$. In this case

$$0 \leq \left(\frac{\partial p}{\partial n}\right) < \rho c \operatorname{div} u$$

can be written by means of

$$\frac{\partial p}{\partial t} = -\rho c^2 \operatorname{div} u \qquad \text{and } p = p_1 + p_2 \text{ as}$$

$$-\frac{\partial p_1}{\partial t} - \frac{\partial p_2}{\partial t} > c\left(\frac{\partial p_1}{\partial n} + \frac{\partial p_2}{\partial n}\right) \geq 0$$

If the normal makes an angle θ with the radius, then

$$\frac{\partial p_1}{\partial n} = \frac{\partial p_1}{\partial r}\cos\theta = -\left(\frac{1}{c}\frac{\partial p_1}{\partial t} + \frac{p_1}{r}\right)\cos\theta$$

Hence the necessary condition for the propagation of a

breaking front is

$$-(1-\cos\theta)\frac{\partial p_1}{\partial t}+\frac{c\,p_1}{r}\cos\theta > \frac{\partial p_2}{\partial t}+c\frac{\partial p_2}{\partial n}$$

From the above it can be shown that a breaking front cannot follow a diverging spherical wave of tension into quiescent liquid where ($p_1 < 0$, $p_2 = 0$), for then we have $\theta = 0$) since over the front $p = p_b = p_1$. In order to cause cavitation, spherical waves of tension must overrun other waves of such character that the right side of the above equation has large negative values much greater than is actually possible.

7. Thermodynamic Considerations

The above theory is based solely upon the laws of conservation of matter and conservation of momentum. The theory reveals, however, that if $p_b < p_c$, then the passage of either a breaking front or a forced closing-front is very likely to leave behind it a greater amount of energy than the kinetic and potential energy of the cavitated liquid can account for. It is very possible, as has been suggested by Smith's theory of pulsating bubbles, that the excess energy is dissipated or radiated away in consequence of the local oscillations associated with the impulsive production and destruction of bubbles.

The difference between p_b and p_c implies an inherent

instability of liquids. There exists no hydrodynamic or thermodynamic reason why cavitation cannot occur at pressures lower than p_c. From the point of view of this theory, this leads to breaking fronts advancing with velocities greater than c, which is very improbable from a region already cavitated. If we attack the problem of cavitation statistically, rather than hydrodynamically, we arrive at relationships which are more readily verifiable by experiment.

STATISTICAL THEORY OF CAVITATION

1. Introduction

A liquid standing at room temperature may be described physically as being in a certain equilibrium state. If the liquid is placed in an ultrasonic field, the equilibrium configuration which the molecules of the liquid possesssed at room temperature is altered until a new equilibrium state is reached. It can be seen that in passing fromone state to another there is a definite rearrangement of the molecules of the liquid. The rate at which this rearrangement proceeds and the new equilibrium configuration which is reached is dependent upon the temperature of the liquid and a characteristic energy of the molecules. It is evident that an initial equilibrium configuration must be assumed. From this initial assumption a statistical theory of cavitation can be developed.

2. Theory of Rate Processes[34]

The Arrhenius equation showing the variation of the specific rate of a physical reaction (such as a rearrangement of the molecules) with temperature can be expressed as

$$k = Ae^{-E/RT}$$

(34) S. Glasstone, K.J.Laidler, H. Eyring, The Theory of Rate Processes, McGraw Hill, New York (1941)

where k = specific rate of the reaction
A = collision number or frequency factor
E = difference in heat content between the activated molecules in the ultrasonic field and the "inert" molecules at room temperature

The quantity E is usually termed the "energy of activation" of a reaction. This represents the energy that a molecule in the "inert" state must acquire from an interaction with the ultrasonic field before it can take part in the rearrangement to the final configuration.

It has been suggested by Sohay(35) that there exists the possibility of a dualism in the realm of sound energy analogous to the dual nature of electromagnetic radiation. If we assume the basis of present quantum theory, it is improbable that a quantum of sound energy does exist in the interaction of sound and matter. This is borne out by the fact that the relatively low frequencies attainable by present day ultrasonic generators would give us energies far below the observable activation energies necessary for a physical reaction to take place. It must be mentioned, however, that this concept has not been fully explored.

A physical reaction is, as has been pointed out, characterized by an initial configuration which passes over by a continuous change of coordinates into a final configuration. "There is, however, always some intermediate configuration which is critical for the process, in the sense that if

(35) B.K.Sohay, Journ. Acoust. Soc. Am. 17, 285 (1946)

this system is attained there is a high probability that the reactions will continue to completion. This critical configuration is called the "activated complex" of the reaction, and is, in general, situated at the highest point of the most favorable reaction path on the potential energy surface."(36) We may regard this activated complex as an ordinary molecule which possesses all the usual thermodynamic properties, except that motion is restricted along the reaction coordinate leading to decomposition at a definite rate. It is now possible to derive the concentration and rate at which these activated complexes pass through the critical configuration of the activated state by statistical methods. The product of the concentration and rate is equal to the specific reaction rate. The important concept to bear in mind at this point is that there exists an activated complex which is the critical intermediate in every rate process.

If we let C' represent the number of activated complexes per unit volume of liquid lying in a length d representing the activated state at the top of a given potential barrier, and $\bar{v}$ the mean velocity of crossing this potential barrier, then we have $\bar{v}/d$ equal to the frequency at which the activated complexes surmount the potential barrier. Hence we have for the rate of reaction

$$\text{Rate of reaction} = C' \frac{\bar{v}}{d}$$

(36) S.Glasstone, et al, p.10, Ref.34

We can treat these activated complexes as ordinary molecules if we write their concentration at the top of a potential barrier as

$$C' = \frac{C\,(2\pi\ m^*\ KT)^{\frac{1}{2}}d}{h}$$

where m* is the effective mass of the activated complex in the coordinate of reaction. The factor $(2\pi\ m^*\ KT)^{\frac{1}{2}}\ d/h$ is the partition function for translation in the reaction path (only possible motion); this partition function represents the probability of the occurrence of the activated complex at the top of the potential barrier. By classical methods it can be shown that

$$\text{Rate of reaction} = C\ \frac{KT}{h}$$

From this it can be derived that if k is the specific reaction rate, then

$$k = \frac{KT}{h}\ K'$$

where K' is the constant for the equilibrium. The constant K' may be related to $\Delta F'$ the standard free energy, $\Delta H'$ the heat content change, and $\Delta S'$ the entropy change by the usual thermodynamic relationships. Hence we may now write the expression for the specific reaction rate for activated complexes surmounting a potential barrier in the form

$$k = \frac{KT}{h}\ e^{-\Delta F'/RT}$$

or

$$k = \frac{KT}{h} e^{-\Delta H'/RT} . e^{\Delta S'/R}$$

It will now be advantageous to transform the equilibrium constant K' and the thermodynamic functions in terms of the partition functions of the species involved by the methods of statistical mechanics. If this is done, we obtain

$$k = \frac{KT}{h} . \frac{F'}{\sum_{i=1}^{N} F_i} e^{-E/RT}$$

for a system of N molecules where F' is the partition function of the activated complex per unit volume, F_i is the partition function of the $i^{\underline{th}}$ reactant in the physical rearrangement and E is the difference at the absolute zero between the energy per mole of activated complex and the sum of the energies of the reactants.

We have, at this point, formulated enough basic theory to explain to a degree the process of ultrasonic cavitation in liquids. It is in order here to assume that the reactants mentioned above are the molecules of a liquid; the potential barrier which is to be surmounted may be explained in terms of the cohesive strength of the liquid; the energy put into the system originates in the ultrasonic transducer; the initial configuration is the "non-cavitated" liquid; and the final configuration is the cavitated liquid.

3. Eyring's Liquid Structure Theory[37]

Eyring regards a liquid as made up of "holes" nearly molecular in size moving about in matter. These holes are considered as playing the same part in a liquid as molecules do in a gas. The holes are assumed to be a part of the natural basic structure of the liquid. This concept differs somewhat from Fürth's theory in that Fürth assumes the natural holes in a liquid to be under high vapour pressure from within, while Eyring's natural cavities are devoid of matter, at least in the initial equilibrium state.

Suppose now we have N molecules which make up a liquid. Each of these N molecules is bound to the others by bonds adding up to the total energy E which is given by

$$E = \sum_i n_i \varepsilon_i$$

where

n_i = number of bonds of the $i^{\underline{th}}$ kind
ε_i = bond strength of the $i^{\underline{th}}$ bond

To vaporize the N molecules requires exactly NE/2 energy, since each bond belongs to two molecules. Providing no hole is left in the liquid upon vaporization, it then takes an amount of energy equal to E/2 to vaporize one molecule. However, if we vaporize one molecule, and leave a hole, we must supply energy equal to E. If we return this molecule to the liquid we bring back energy equal to E/2. It is seen then that it requires the same energy E/2 to make a hole in

(37) H.Eyring, Ref.25

a liquid of molecular size as it does to vaporize a molecule without leaving a hole.

If we consider a molecule in empty space it is evident that it has a higher entropy than when in a liquid, since it has a large number of accessible states without increase in energy. The same is true of the hole dissolved in the liquid. Now a hole can have many positions in a liquid and still have the same energy. These holes, like molecules, can be pictured as having a random motion with a velocity characteristic of the molecules at that temperature. A liquid can be thought of as having a definite lattice arrangement somewhat like a crystal. Wherever a hole exists, a molecule close by can jump into an empty lattice point, which leaves a hole behind, and as this process goes on continuously each hole contributes a new degree of translation to the liquid. If the liquid is at a temperature below the critical or boiling temperature, this translation in the liquid is not as free as in the vapour phase, hence the partition function for holes is smaller than for vapour molecules. Also, below this critical temperature a molecule in going from one hole to another has to surmount the potential barrier mentioned above. At the critical temperature the mobilities and partition functions of the holes and the molecules are equal, hence for holes in liquids or molecules in empty space we have the theorem of equal entropies and energies. It is intuitive

that the energy required for the association or dissociation of holes of molecular size depends primarily upon the nature of the liquid.

Since the physical process of cavitation takes place in both the liquid and gas phase, we assume for a first approximation that association or dissociation does not affect the distribution ratio of holes between the liquid and gas phase. If this is assumed, we can say that at the critical temperature we should have as much matter in a unit volume of otherwise empty space as we have holes in a unit volume of matter. As the temperature is lowered, a plot of the average density of the liquid as a function of temperature would almost parallel the temperature axis, the density increasing slightly as the temperature decreases. Hence the average density of the vapour and liquid phase should be nearly independent of the vapour pressure or the temperature. "Now the law of rectilinear diameter states that for actual cases such a plot of average density is a straight line, but that it may, and in general is, a little inclined to the temperature axis. Thus the law of Cailletet and Mathias follows from statistical mechanical considerations of holes."(38)

The theorem of equal energy and nearly equal entropies for holes in matter as for molecules in empty space frequently applies in the same way to the solid state as to the liquid state. We can surmise, therefore, that the vapour is a rough

(38) H.Eyring, P.285, Ref.25

measurement of the number of holes in the condensed phase.

The theory of holes is of importance in all phenomena in which a part of the matter in a condensed phase moves with respect to other parts. This follows from the fact that at the borders of holes such motions can be set up with a minimum of disturbance to molecules not participating in the motion.

In certain phenomena it may be much simpler to fix attention on the few holes rather than on the much more numerous molecules. Only in very precisely ordered arrangements, can the attainment of a critical motion by a few molecules profoundly affect the stability of neighbouring molecules and these in turn release still others from their restraining potential.

Some support for the general point of view with respect to holes as developed here may be obtained from discussions of the Wilson theory of semiconductors. In this theory, electrical conductance is assumed to arise from thermal fluctuations which raises an electron of low energy up to the conduction levels leaving a positive hole behind. This theory seems to account satisfactorily for the observed facts.(39)

4. Interpretation of Cavitation and Conclusions

It was mentioned earlier, in conjunction with Briggs'

(39) A.Fowler, Proc.Roy.Soc. A140, 505 (1933)

experiments on the cavitation strength of liquids, that the only property of a liquid consistently related to the cohesive pressure was its viscosity. An extension of Eyring's[40] theory of absolute reaction rates to viscosity, coupled with his "theory of holes" as applied to liquid structure, yields some information concerning this relationship.

If we consider two layers of molecules a distance δ_1 apart, with a force F_0 tending to displace one layer with respect to the other, then the coefficient of viscosity is given by

$$\eta = \frac{F_0 \delta_1}{\Delta v}$$

where v is the difference in the velocity of the two layers. The motion of a layer of molecules is assumed to involve passage of a molecule from one equilibrium position to another. For this passage to occur, it is required that a suitable hole be available, and the production of such a hole requires energy. Hence the passage of a molecule from one equilibrium position to another is the same as the system surmounting a potential barrier. In order to determine the energy involved, it is convenient to adopt the following notation. Let δ be the distance between two equilibrium positions and δ_3 the distance between two molecules, both measured in the direction of motion. Also let δ_2 be the mean distance between two adjacent molecules in a direction at right angles to the direction of motion. If we assume a symmetrical

(40) S. Glasstone, et al., p.480, Ref. 34

potential energy barrier, then the distance between the equilibrium position and the activated state (at the top of the barrier) is equal to $\frac{1}{2}\delta$. The energy acquired by a moving molecule when it has reached the top of the potential barrier is then given by $\frac{1}{2} F_o$.

With the assumption that the transmission coefficient for the passage of a molecule over a potential energy barrier will be unity and neglect tunneling through the barrier, then the rate of passage of a molecule over the barrier is

$$k = \frac{KT}{h} \frac{F'}{F} e^{-E/RT}$$

when no force is applied. According to Briggs[41], the forward reaction rate k is given by

$$k_f = k\, e^{(F_o \delta \delta_2 \delta_3)/2KT}$$

while the reaction rate for a molecule that has once surmounted a potential barrier to return to its initial state may be stated as

$$k_b = k\, e^{-(F_o \delta \delta_2 \delta_3)/2KT}$$

Then Δv for each molecule in the faster layer is

$$\Delta v = \delta k \left[e^{(F_o \delta \delta_2 \delta_3)/2KT} - e^{-(F_o \delta \delta_2 \delta_3)/2KT} \right]$$

or

$$\Delta v = 2 \delta k \sinh (F_o \delta \delta_2 \delta_3)/2KT$$

(41) H.B.Briggs, et al, p.674, Ref. 13

With this equation for Δv we may write the expression for the viscosity as

$$\eta = F_0 \delta_1 / 2\delta k \sinh (F_0 \delta \delta_2 \delta_3)/2KT$$

or

$$\eta \simeq \delta_1 KT / \delta^2 \delta_2 \delta_3 k$$

if we assume that $F_0 \delta \delta_2 \delta_3 << 2KT$, as is the case for very small forces. If we introduce the value of k and δ_1 assume δ equal to δ_1, we get for the viscosity

$$\eta = \frac{N h e^{E/KT}}{V(1 - e^{-h\nu_i/KT})}$$

or

$$\eta = A e^{E/KT}$$

where N is Avogadro's number and V the molal volume of the liquid which may be taken on the average to be about 40 cm^3. In order to arrive at the above expression for the viscosity, Eyring assumes that

$$\frac{F'}{F} = \left(1 - e^{-h\nu_i/KT}\right)$$

where ν_i are frequencies, having a range between 1.4 and 1.8×10^{12}, which can be obtained from the specific heats of the liquids. With this range we get F'/F values between 0.1 and 0.25. From the average value of V we see that the factor Nh/V is of the order of 10^{-4}. This must be multiplied by a factor 4 to 10 to account for F/F'. Hence the constant A in the last equation for viscosity varies between 4×10^{-4}

and 10^{-3}, depending on the liquid. We must now consider the activation energy, E, which is related to the internal energy plus the energy necessary to overcome the external pressure.

Eyring assumes that following van der Waals' equation

$$(p + a/v^2)\,(v - b) = RT$$

We may write for the activation energy

$$E = (p + a/v^2)\mathcal{S}$$

where $\mathcal{S}$ is a volume equal to about 2v. If this is substituted in our last expression for the viscosity, we get

$$\eta = A \exp\left[\frac{(p + a/v^2)}{RT}\right]$$

When the pressure, p, is zero then

$$E = (a/v^2)\,(\mathcal{S}/N)$$

It is clear that as p increases E increases and hence the viscosity should increase. If we have a negative pressure, which is always the case when cavitation occurs, the energy will decrease, and if $p = -a/v^2$ the activation energy should decrease to zero and the potential barriers disappear. At this point the liquid becomes gas-like; p cannot then be more negative than a/v^2, the van der Waals cohesive pressure. As was mentioned earlier, a negative pressure of about 10^4

atmospheres fulfills this limiting condition. This is about of the same order of magnitude as the theoretical cohesive pressure for a liquid.

However, by similar considerations for solids the actual strength has been found experimentally to be only 1/1000 of the theoretical values. Then for liquids we should expect to have to multiply p by a factor β which is of the order of 1000 when the applied pressure is negative. If, however, the pressure is positive, then it is very probable that the factor β is close to unity in order to get the observed dependence of viscosity and pressure. Since in an ultrasonic field the pressure varies with time from positive to negative values, we should expect β to be a function of time. Then we can write

$$\eta = A \exp\left(\frac{(\beta p + a/v^2)/S}{RT} \right)$$

where β is a function of time and the ratio p/p_c (p_c being the cohesive pressure of the liquid) is equal to 1000 for steady state negative pressure nearly equal to the cohesive pressure, and also equal to unity for positive pressures.

If we assume the theoretical derivation of these equations, then we can interpret some of the experimental results obtained by Briggs.[42] In Fig. 4 we have plotted the cavitation for degassed kerosene. The value of a/v^2 for this liquid is small

(42) H.B.Briggs, et al, p.675, Ref. 13

but positive, and it was found that it requires a negative pressure of about 0.8 to 1 atmosphere to cause cavitation. With the application of a hydrostatic pressure we change a/v^2 but little, and hence it requires the same negative pressure to cause cavitation. However, since we have introduced a hydrostatic pressure of A atmospheres, it requires an acoustic pressure of A + 1 atmospheres to produce a negative pressure in the liquid of one atmosphere. We obtain, therefore, a straight line relationship between the amplitude of the sound pressure and the hydrostatic pressure of the liquid. We assume here that cavitation input voltage is a function of the sound pressure. The straight line obtained is displaced from the zero hydrostatic pressure origin by the amount of the cohesive pressure of the liquid.

The last equation for the viscosity of a liquid, derived above, explains the dependence of cavitation pressure on viscosity observed in Fig. 5 and given by the equation

$$\ln(\eta/\eta_0) = 3.05\, p_c$$

If we assume that

$$A = \frac{N h}{V\left(1 - e^{-h\nu_i/KT}\right)}$$

is equivalent to η_0 above, then

$$\ln(\eta/A) = \alpha S/RT v^2 = E/KT$$

where η_0 is fairly constant from liquid to liquid.

Also if we assume that the value of β is constant from

liquid to liquid, we should have cavitation occurring at a negative pressure equal to

$$-p_c = a/\beta v^2$$

Hence we may now evaluate the constant β since

$$\ln h/A = \frac{3.05\ p_c}{10^6}$$

$$\ln h/A = \left(\frac{3.05}{10^6}\ \frac{a/v^2}{\beta}\right)$$

$$\ln h/A = \frac{(a/v^2)\ S}{RT}$$

or

$$\beta = \frac{(3.05)\ RT}{10^6\ S}$$

if p_c is given in dynes/cm^2. Now RT at 300°K is equal to 2.42×10^{10}, and according to Eyring S is about $2v = 80$ cm^3. Hence β is about 930, which is in good agreement with experimental observations.

The curve of Fig. 6 indicates that the constant β is a function of time. If we take

$$\beta = 930\left(1 - e^{-\alpha t^{1/3}}\right) + 1$$

where α is about 2.2 for castor oil, then the experimental curve is fitted within the accuracy of measurement. It can be assumed that the above equation holds down to very short time

intervals; then the time necessary for β to double is given by

$$t_o^{1/3} = 1/(930)(2.2)$$

or

$$t_o = 1.2 \times 10^{-10} \text{sec.}$$

which is the time required for a force of half the theoretical breaking strength to produce a cavity in the liquid. "This should probably occur when the cavity size has grown from 1 to 2 units, and hence should compare with the transition time for a molecule jumping from one hole to another."(43) Eyring has calculated this time to be

$$t = \frac{1}{k} = \left(\frac{h}{KT}\right)\left(\frac{F}{F'}\right) e^{E/KT}$$

or

$$t \simeq 1.6 \times 10^{-12}\, e^{E/KT} \text{ sec}$$

For castor oil upon which no external force is acting, we get from activation energy measurements a value for E/KT equal to 8.6. Now for a negative pressure of half the breaking strength this factor is reduced to 4.3 and hence the time should be

$$t_o = 1.6 \times 10^{-12} e^{-4.3} = 1.15 \times 10^{-10} \text{ sec.}$$

which is in good agreement with the value of 1.2×10^{-10} sec. found experimentally.

(43) H.B.Briggs, et al, Ref. 42

It would seem that the more recent experimental evidence is in favor of Eyring's statistical theory of cavitation, and that a thorough hydrodynamic theory becomes too complex to be interpreted and tested experimentally.

ABSTRACT

The input of ultrasonic power into a liquid medium causes a definite rearrangement of the initial molecular configuration. When cavitation occurs, it is observed that small cavities, filled with the vapour of the liquid, are formed in the liquid. These cavities vanish as soon as the ultrasonic power source is shut off, and the liquid presumably returns to its original state. It is necessary, therefore, to assume an initial molecular configuration, i.e., postulate a basic liquid structure theory, the liquid being under standard conditions of pressure and temperature. Further, a "transition theory" describing the passage of the initial molecular configuration to the final cavitated configuration must be worked out. Cavitation in a gassy and a completely degassed liquid would then seem to require two separate theories for its occurrence, since their initial molecular configurations would appear to differ widely. This difficulty can be surmounted, however, if we assume that a completely degassed liquid is made up of natural holes which are molecular in size and even, perhaps, the size of two or three molecules. Holes larger than the size of a molecule may be formed by the combination of several unit size holes. This produces a metastable condition, unless the liquid happens

to be in a force field which would activate and sustain such a chain of events. Hence, an ultrasonic field gives us a source for the required "activation energy", as well as a source of "sustaining energy" resulting in further combination of the "nuclei" cavities until a definite break or fracture occurs. Although this point of view can be applied strictly to degassed liquids, it is probable that a completely degassed liquid does not exist. Hence, it would appear more tenable to assume that both natural holes, as well as microscopic gas bubbles, exist in liquids under standard conditions. Although at the initial cavitation point the natural holes and the gas bubbles behave differently, it is probably a very short time before the cavities which cause the fracture of the liquid consist of gas bubbles alone. These are the visible cavities observed in cavitation experiments.

It has been observed that cavitation takes place during the negative half of the pressure cycle of an ultrasonic field. If we assume that a certain activation energy must be supplied to a molecule to cause it to alter the initial configuration, then it is reasonable to conclude that the effect of a negative pressure is to decrease the potential energy barrier, making it easier for a molecule to "jump" away from a hole. This causes the cavities to increase in size. The probability that the effect of a negative pressure is to increase the tunneling through the potential can be taken as

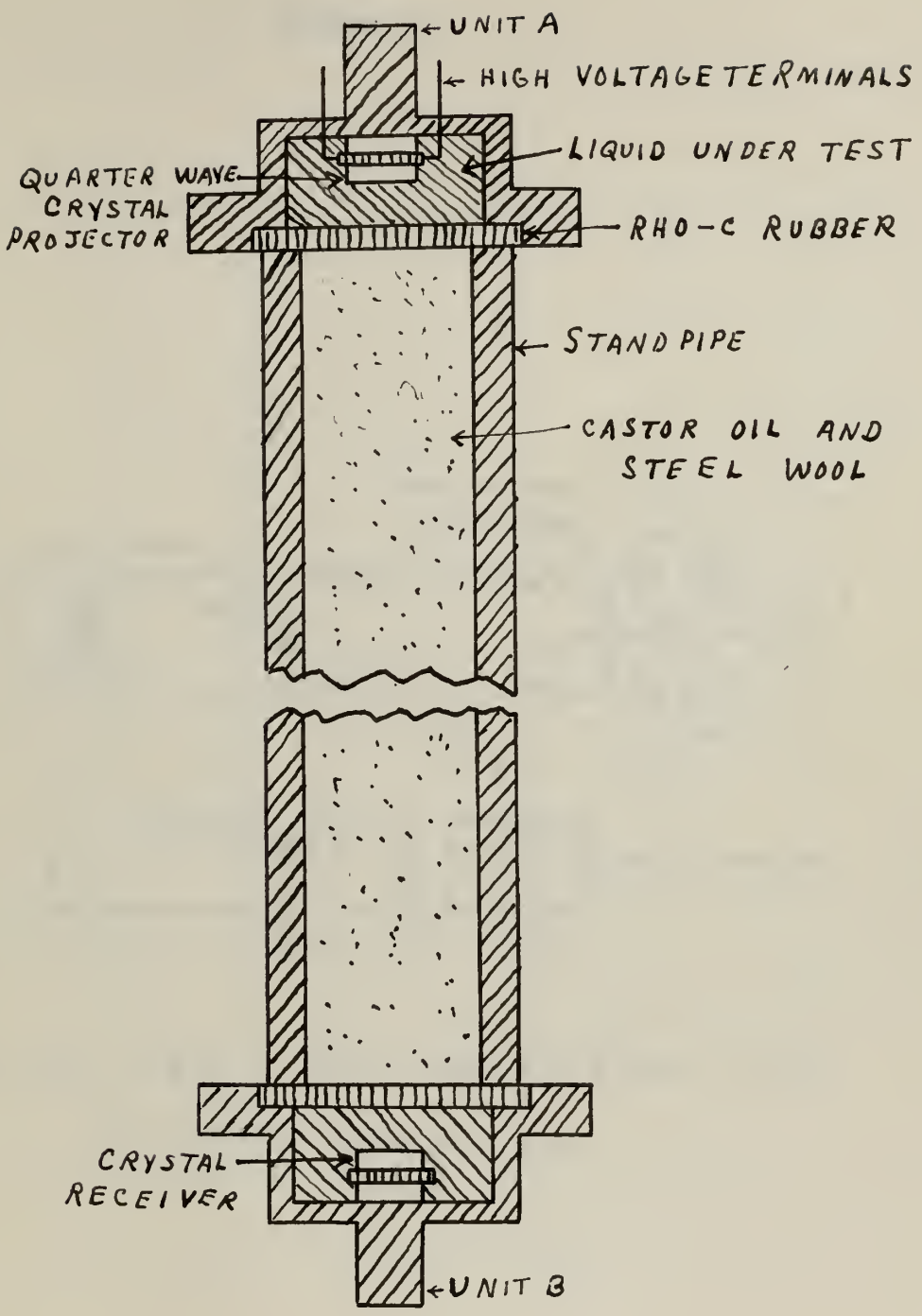
being too low to have any effect. Conversely, the effect of a positive pressure is to increase the potential barrier for a molecule to jump away from a hole, tending to decrease the size of the existing cavities.

If the above reasoning is followed, it appears that a reversible process is occurring, i.e., we have the formation of cavities during the negative half of the pressure cycle, and the destruction of cavities during the positive half of the cycle. This must be true so long as the sound pressure does not exceed the cohesive pressure of the liquid. However, once the cohesive pressure is exceeded the reversibility ceases, with the result that cavities are formed and increase in size until there is complete rupture of the liquid. There is also the possibility that the existence of microscopic bubbles in the liquid may cause the reversibility to shift from equilibrium at some point before the sound pressure exceeds the cohesive pressure of the liquid. Recent experiments with liquids known to contain these gas bubbles seem to indicate that this is the case.

The relationship between the activation energy and the cohesive strength of a liquid has been determined. From this, it can be concluded that the statistical theory of cavitation, as suggested by Eyring's theory of physical processes in liquids, is the more tenable theory of cavitation, and offers the better avenue for further investigation.

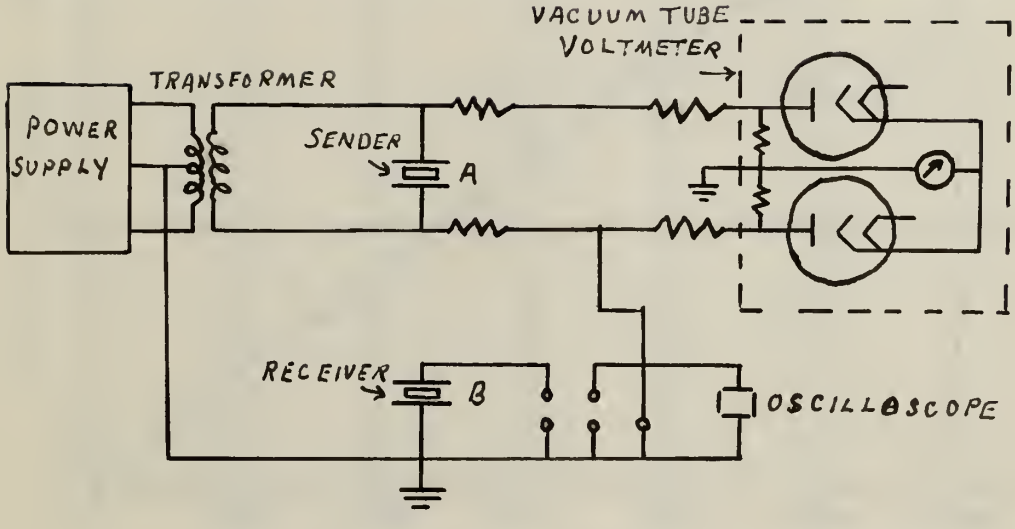
ILLUSTRATIONS

Figure 1



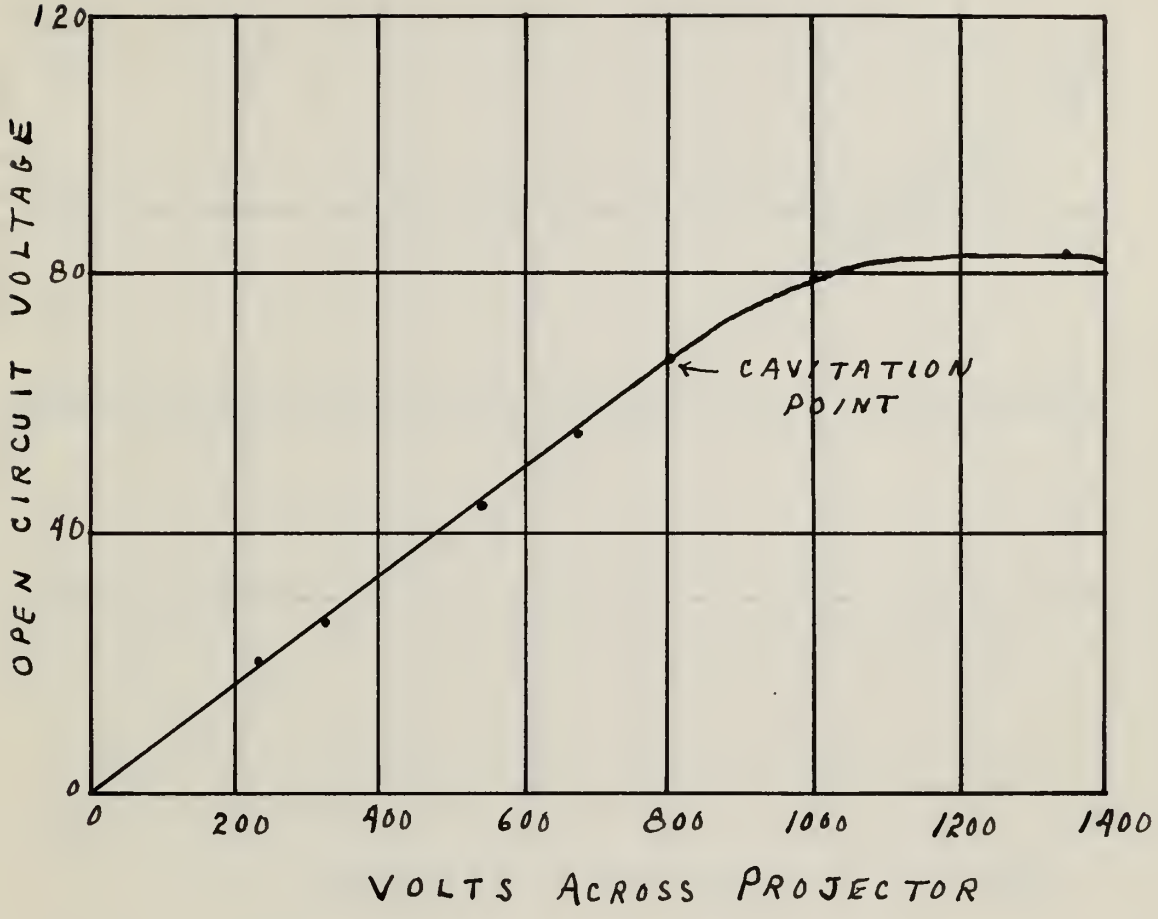
CRYSTAL UNITS AND EXPERIMENTAL STANDPIPE FOR STUDYING CAVITATION.

Figure 2



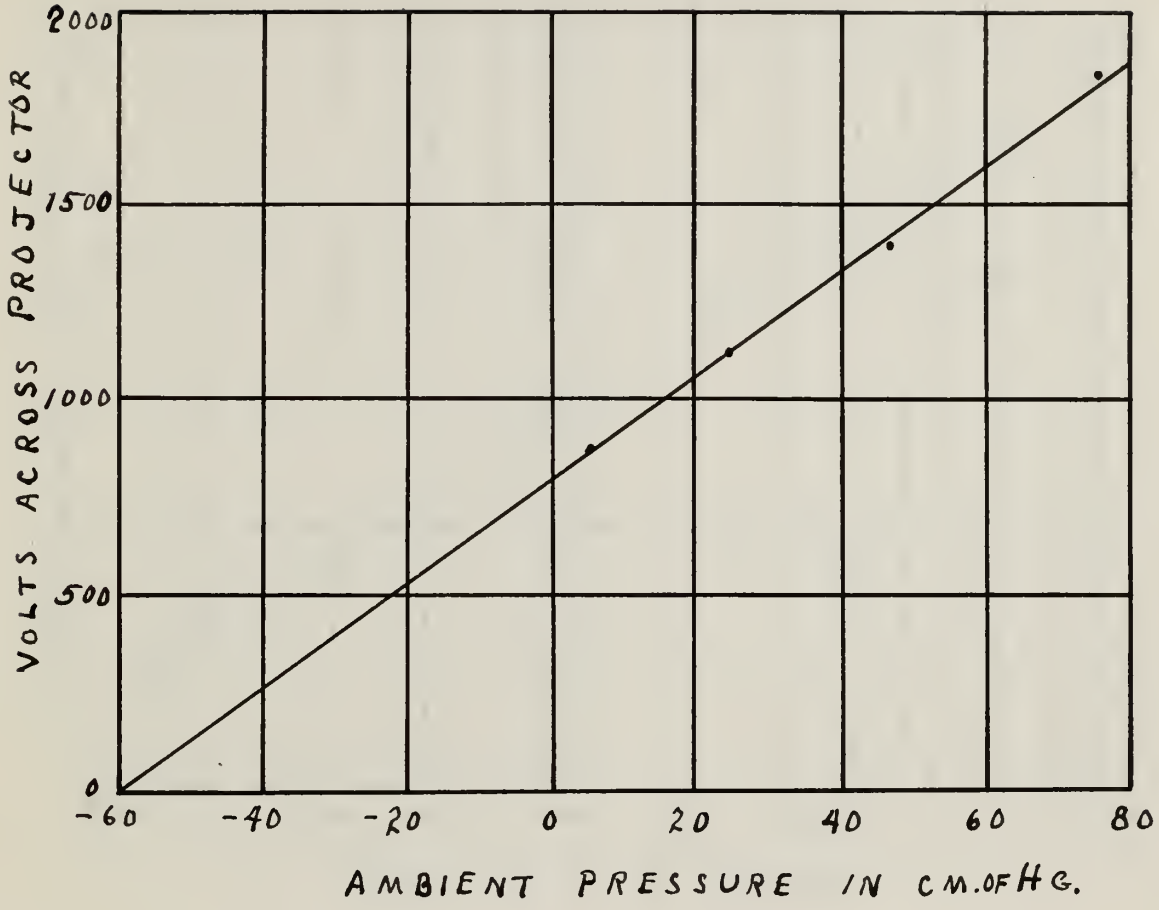
PULSING CIRCUIT AND OSCILLOGRAPH FOR STUDYING CAVITATION

Figure 3



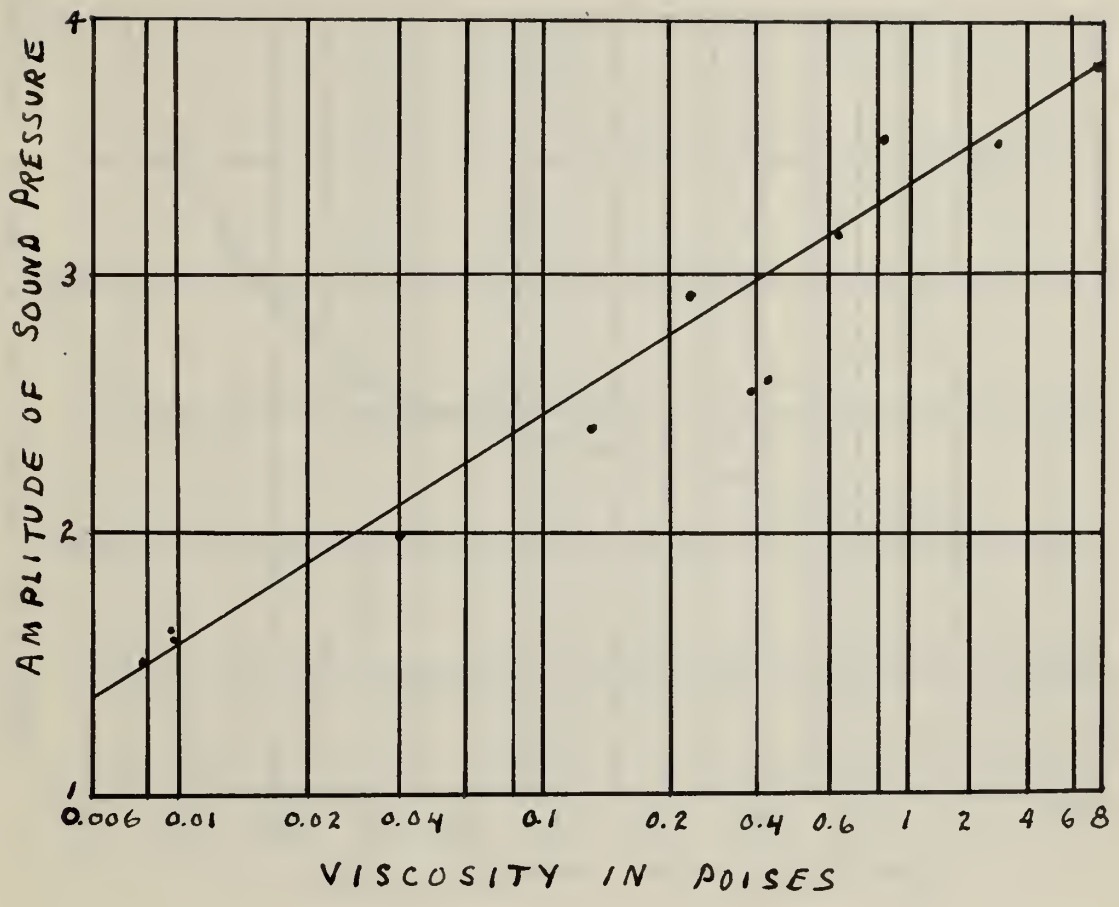
INPUT VERSUS OUTPUT VOLTAGE FOR NON-DEGASSED KEROSENE

Figure 4



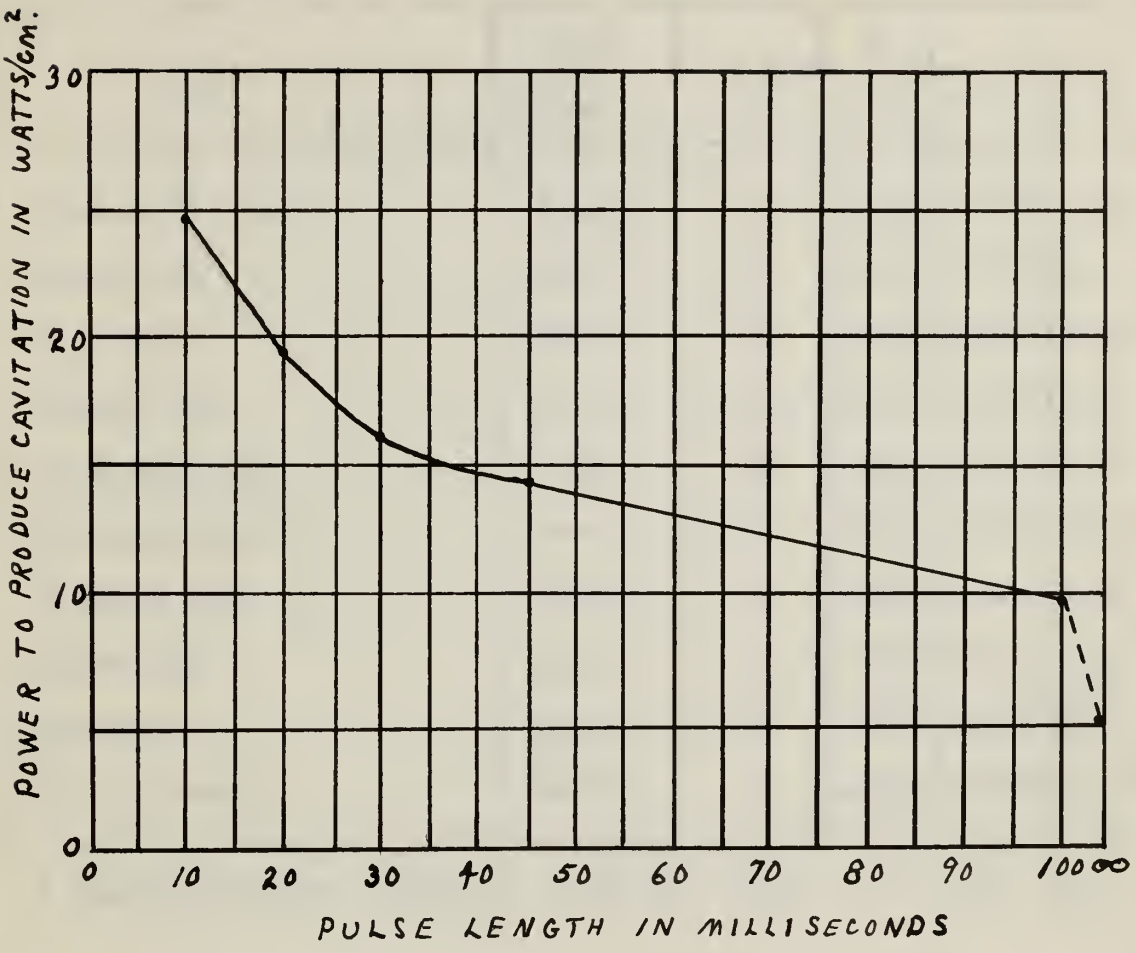
CAVITATION INPUT VOLTAGE PLOTTED AGAINST AMBIENT PRESSURE FOR DEGASSED KEROSENE

Figure 5



AMPLITUDE OF CAVITATION SOUND PRESSURE PLOTTED AGAINST VISCOSITY OF THE LIQUID.

Figure 6



CAVITATION SOUND PRESSURE PLOTTED AGAINST PULSE LENGTH FOR DEGASSED CASTOR OIL.

TABLE I

Name	Visc. in poises	V.P.*	$\nu \times 10^5$	ρ	ρ_c
Castor Oil D.B.	6.30	A	1.477	0.969	3.90
Olive Oil	0.84	B	1.431	0.912	3.61
Corn Oil	0.63	B	1.463	0.914	3.05
Peanut Oil	----	B	1.458	0.936	3.08
Soya Bean Oil	0.45	B	1.461	0.919	2.45
Cottonseed Oil	----	B	1.463	0.912	2.32
Linseed Oil	0.38	B	1.468	0.921	2.36
Sperm Oil	0.25	B	1.440	0.880	2.90
Kerosene	0.04	B	1.324	0.810	2.00
G. E. Transil	0.13	B	1.350	0.880	2.35

* Vapor pressure ranges A 0.01 cm. Hg., B 0.1 cm. Hg.

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